Nuclear Spins of Silver-104 and Silver-106*

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The nuclear spins of four neutron-deficient isotopes of silver have been measured by atomic-beam methods. The results are: for 27-min Ag^{104}, $I=2$; for 1.2-hr Ag^{104}, $I=5$; for 24-min Ag^{106}, $I=1$; and for 8.3-day Ag^{106}, $I=6$. Of these, the result for 1.2-hr Ag^{104} has not been reported previously.

INTRODUCTION

THE use of an auxiliary evaporator has made possible the unambiguous determination of the nuclear spins of several neutron-deficient silver isotopes.[1] The spin measurements were made with an atomic beam magnetic resonance apparatus,[2] operating on the flop-in resonance principle.[3] The beam was detected by its radioactivity, and the decay curve of each sample was analyzed to determine its composition. While the evaporator method outlined below could be used for measurements[4] on 40-day Ag^{105} and 8.3-day Ag^{106}, this has not been done. Rather, the shorter-lived ($T_{\frac{1}{2}} \leqslant 1$ hr) isotopes have been investigated, and the longer activities have been treated as a background of comparatively long half-life. This work roughly parallels that recently performed at Princeton[5] except with respect to the method of isotope production.

ISOTOPE PRODUCTION

Neutron-deficient isotopes are produced in the Crocker 60-in. cyclotron on the Berkeley campus of the University of California. This cyclotron delivers alpha particles, deuterons, or protons with an energy of nearly 12 Mev/nucleon in an external beam. Using a Pd target, the proton or deuteron beam is of sufficient energy to produce those silver isotopes having $A \geqslant 104$ (the restriction is due to the low abundance of Pd^{102}). The $Pd(p,n)Ag$ reaction has been used[4] successfully to produce the isotopes with long half-lives and also the 24- and 27-minute pair. These two, however, present a problem in identification. The half-lives are too nearly alike to get a definite isotope assignment from the decay of a given spin sample. To obviate this difficulty, the (α,kn) reaction on rhodium was used to make positive spin assignments. By suitably degrading the energy of the alpha beam, any reaction for which $k \leqslant 4$ can be made the dominant one. This means that any one of the isotopes or isomeric pairs for which $103 \leqslant A \leqslant 106$ can be produced preferentially. To determine the degradation of the beam that is necessary to produce isotopes of each value of A, a "stacked-foil" experiment was performed on rhodium. In this experiment, 12 1-mil Rh foils were marked for identification and stacked in a target holder such that the alpha beam could not enter a given foil until it had penetrated all the preceding foils. After a cyclotron exposure to an integrated beam of about 10–15 μa-sec, the target was quickly broken open and the foils separated. Each foil was mounted for ease of handling on a standard beam-collecting "button." The buttons were then rotated through four crystal counters and two continuous-flow gas β counters. Two of the crystal counters were set to count the silver x-rays, while the other two were set to count higher energy radiation $\geqslant 100$ kev. (The crystals are too thin to provide appreciable resolution for γ rays in this energy range, but they do function with poor resolution and reduced efficiency.) The time required for a complete cycle, with 1-min counts in each counter, was ~ 20 min. This cycle time was sufficiently short that quite reliable decay curves were obtained, even for the half-hour activities. After the first cycle, it was clear that 10 mils of rhodium had stopped almost all the alpha beam. Therefore the decay of only the first ten foil samples was studied. The decay curve for each of these samples was then analyzed into two components: $T_{\frac{1}{2}} \sim 1$ hr (Ag^{103},Ag^{104}) and $T_{\frac{1}{2}} \sim \frac{1}{2}$ hr (Ag^{104},Ag^{106}). The results of this analysis for the data taken in the crystal counters set to count high-energy radiation are shown in Fig. 1. The peaks of each curve are readily explained from known isotope data[6–9] and are noted in the figure. This experiment gives enough information that it is possible to produce a silver isotope of given A with a minimum of contamination from adjacent isotopes. Proper degradation of the alpha beam thus provides a sharp separation of the two isotopes (104, 106) having approximately 30-min half-lives. Either of the isomeric pairs at these values of A can be produced as desired. The same should be true for

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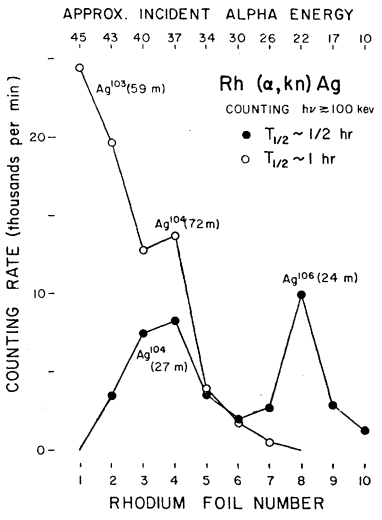

FIG. 1. Stacked-foil experiment, showing the relative amounts of ~30-min and ~60-min activity found on each of the ten 1-mil rhodium foils. Only nuclear gamma transitions of energy ≥100 kev were counted. The upper scale gives the approximate energy of the alpha beam as it entered each foil.

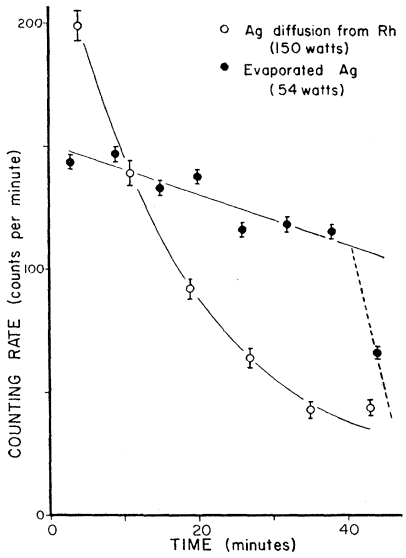

FIG. 2. Comparison of beam stability (a) when the silver is diffusing directly out of the rhodium target, and (b) when the silver has previously been physically separated from the rhodium. All points have been corrected for the radioactive decay of the beam.

$A=105$, although no attempt was made here because a short-lived isomer has not been observed, while the 40-day Ag^{105} is more conveniently produced from Pd. The available alpha energy is too low to give a complete separation of Ag^{103} from the Ag^{104} pair, but the lighter isotope can be produced. Its nonintegral spin prevents any confusion with the 104-isotope of similar half-life.

ISOTOPE SEPARATION

Although some satisfactory results have been obtained by boiling the active silver directly out of the target palladium or rhodium,[4,5] it is usually more desirable to have the silver separated from the target material before a run is made. The principal advantage is the increased control over and stability of the beam (Fig. 2). A secondary consideration is the lower temperature that can be used with the pure silver and the consequent decrease in problems of heat dissipation.

The chemical separation of silver from palladium is straightforward.[4] This chemistry has been performed in a time short enough that a resonance of 24-min Ag^{106} could be obtained. For the rhodium targets, a different procedure had to be employed, since rhodium is almost insoluble in the standard reagents. A method that couples speed with convenience has been used.

In this method, the irradiated rhodium foil is placed in a tantalum "pot" which has a collimating snout in the lid. This pot is placed on pins in a vacuum and heated by electron bombardment until the rhodium melts. At the pressures used, the boiling temperature of silver is lower than the melting temperature of rhodium. Therefore, when the rhodium melts, the radioactive silver is released and escapes through the collimating snout. The snout is directed into a standard atomic beam oven, which is cooled by contact with water-cooled copper surfaces. After this evaporation process the oven is capped, and the experiment is performed in the usual way. A controlled amount of stable silver is evaporated previously to act as a carrier for the radioactive beam. The minimum time required for this physical separation is about one hour. Since the half-lives of the shortest activities that are of interest here are about $\frac{1}{2}$ hr, a high level of activity is necessary. A typical oven load may read 50 r/hr at 8 in. before being capped. Most of the radiation is easily absorbed, however, and closing the oven lowers the level considerably. The most satisfactory runs were made with a monitored radioactive beam of 3000–4000 counts/minute per minute of exposure. If the separation has been satisfactory, this level holds almost steady for about $\frac{1}{2}$ hr, then falls suddenly as the oven is emptied. (Figure 2.)

EXPERIMENT

The standard atomic-beam spin determination experiment consists of a low-field measurement of the splitting of the upper angular momentum state ($F=I+J$). In the Zeeman (low-field) region, where nuclear (I) and electronic (J) angular momenta are not decoupled, this splitting is proportional to the magnetic field, the electronic g factor, and the quantity $1/2F$ (for $J=\frac{1}{2}$; $F=I+\frac{1}{2}$). If the electronic g factor is known approximately, then a knowledge of the transition field strength makes the splitting of the energy levels a function only of nuclear spin. In particular, corresponding to the discrete values of nuclear spin that are possible, there is a set of discrete energy differences, $\Delta W(I)$. If these energy differences are supplied successively by an rf oscillator, the probability of transitions among the energy levels is a maximum when the oscillator frequency satisfies the condition

$$\nu=\Delta W(I)/h.$$

Therefore the nuclear spin can be determined by a (usually) small number of tests for the occurrence of transitions among the levels as split by the small applied magnetic field. The atomic-beam apparatus makes possible the performance of these tests on a beam of neutral atoms. In the flop-in apparatus, a transition in the uniform field region between two Stern-Gerlach deflecting fields is necessary in order to satisfy the focussing conditions of the apparatus. Thus, the occurrence of such transitions is detected by an increase in the number of atoms that pass through the apparatus and reach the detector. If the atoms are radioactive, they may be collected on some surface, where they can then be counted. For the silver atoms, a satisfactory collecting surface is fused sulfur. The sulfur is mounted in a brass "button" for ease of handling. The buttons are designed to place the sulfur surface in a precisely reproducible position, (a) with respect to the beam in the apparatus, and (b) with respect to the sodium-iodide crystal or counting chamber in the counters.

At the oven temperatures necessary to maintain a beam of silver atoms, a substantial fraction of the atoms have velocities so large that they can pass through the magnetic region of the apparatus without suffering a deflection large enough to cause them to miss the detector. In practice, this fraction may approach 50% of the radioactive beam. The fact that a fraction of the active beam can be detected in spite of the magnetic fields makes it possible to monitor the beam without disturbing the apparatus by turning off the deflecting fields. During exposures at possible resonance frequencies, where a very low signal must be identified, the fast fraction is cut out by a "stop wire" placed on the axis of the apparatus. This serves to stop all atoms that do not receive at least a certain minimum deflection, i.e., those atoms for most of which it is not possible to detect the desired rf transition.

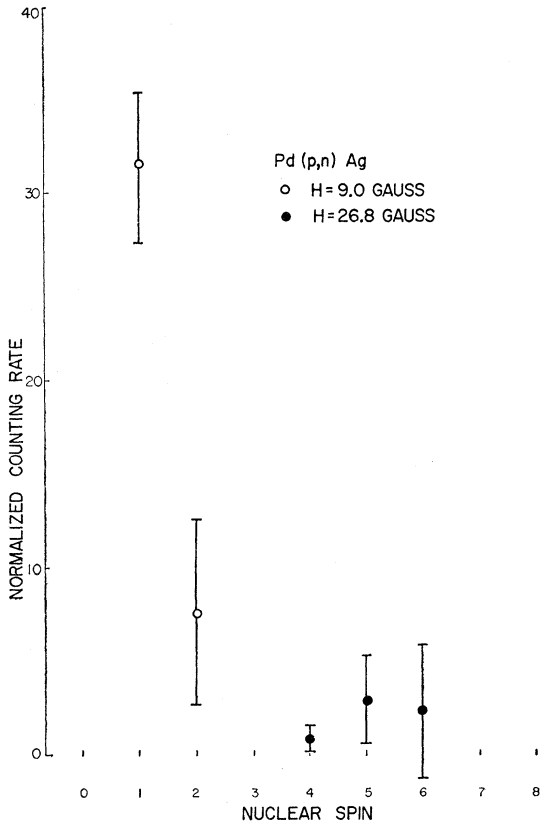

FIG. 4. Spin search for short-lived silver isotopes produced by the reaction Pd(p,n)Ag. The exposures for high spins were made at a higher field to prevent overlap of resonances.

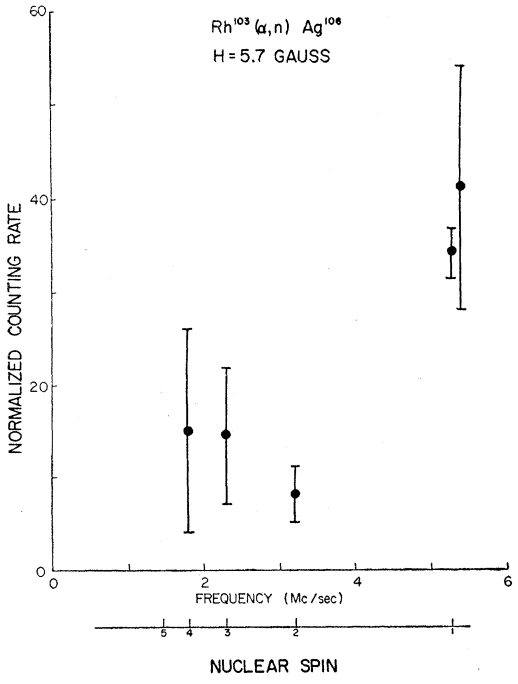

FIG. 3. Spin search for 24-min Ag^{106} at 5.7 gauss.

After the collector buttons have been exposed at the frequencies corresponding to the possible spin resonances, they are cycled through the counters as many times as are warranted by the counting rates. The resulting decay curves are analyzed by an IBM 650 computer, which fits by least squares as many as five components of known half-lives and unknown amplitudes. Because of the low counting rates and the necessarily short counting periods, it is not normally possible to obtain a clear separation of two isotopes whose half-lives are nearly the same. In the described experiments, a maximum amount of information was usually obtained by analyzing the decay curves as being made up of two components, having half-lives of one hour and one-half hour. Separation of the shorter-lived isotopes is made absolutely by a knowledge of the dominant cyclotron reaction. The 1.2-hr Ag^{104} can be made the dominant reaction by slightly degrading the alpha beam. Furthermore, the 59-min Ag^{103} cannot have

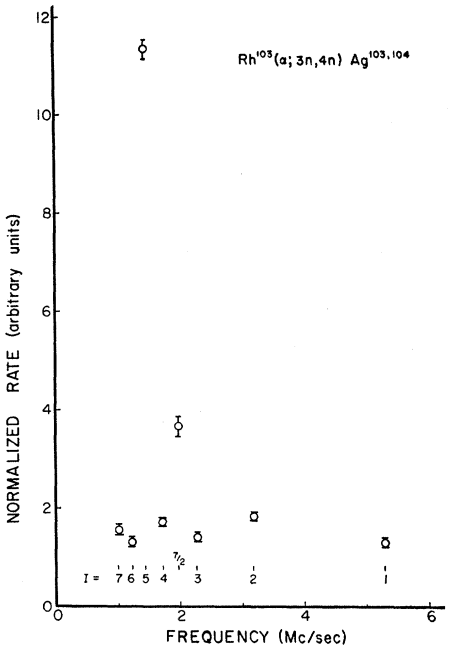

FIG. 5. Spin search for ~1-hr silver activities. The exposure on $I=\frac{7}{2}$ shows the separation of the two resonances, for Ag^{103} and Ag^{104}.

an integral spin. There is thus almost no possibility for confusion in spin assignments.

Since the atomic beam in the apparatus is not exactly steady during the experiment, it is necessary to apply some kind of normalization to the counting rates as they emerge from the counters. This is provided by periodic sampling of the fast fraction of the radioactive beam. The counting rates of these so-called "half-beam" samples are plotted as a function of time. Usually, these points can be averaged by a smooth curve, from which a number proportional to the beam intensity can be obtained for any time at which a spin sample was exposed. After this normalization, an examination of the results will show which spin sample contains an enrichment of each isotope.

RESULTS

The experimental results leading to the assignment of $I=6$ to 8.3-day Ag^{106} have been described previously.[4] At that time the signal corresponding to $I=6$ had been observed at several frequencies from 2.4 Mc/sec to 15.7 Mc/sec. No further attempt has been made to identify the isotope more positively.

The short-lived Ag^{106} isomer has been made from rhodium by degrading the alpha beam to ~23 Mev. This has been accomplished by placing a 21-mil thickness of aluminum on top of 1- or 2-mil rhodium foil. The decay of a sample of silver extracted from such a rhodium target shows almost pure 24-min Ag^{106}. With essentially only one isotope present, the spin identification must be by a comparison of normalized counting rates of samples exposed at frequencies corresponding to various integral spins. Such a comparison is made graphically in Fig. 3. The two exposures corresponding to $I=1$ were made before and after the rest of the spin search. Both show a higher normalized counting rate than the other possible spin values, thus verifying $I=1$ for 24-min Ag^{106}. A signal with ~$\frac{1}{2}$-hr half-life has also been observed, using the (p,n) reaction on palladium (Fig. 4). The low abundance of Pd^{104} would suggest that this signal was produced by Ag^{106}, but the rhodium bombardment provides conclusive evidence.

The 1.2-hr isomer of Ag^{104} is produced in abundance by the $(\alpha,3n)$ reaction on rhodium. A cover foil of 5- to 7-mil aluminum on the 3- or 4-mil rhodium foil degrades the alpha beam sufficiently to peak the $(\alpha,3n)$ reaction in the foil (Fig. 1). The ~1-hr signal obtained for a high spin was at first confused with the $I=\frac{7}{2}$ of Ag^{103}, but more precise measurements at a higher field clearly separated the two resonances. A complete spin search at 5.7 gauss is shown in Fig. 5. A sample was also exposed for $I=\frac{7}{2}$ to illustrate the separation. The availability of comparatively large quantities of 1.2-hr Ag^{104} isotope (even after allowing the 27-min isomer to decay) makes it one of the easiest short-lived silvers with which to work. The reduced size of the $I=\frac{7}{2}$ signal is presumed to be due to the difficulty of causing

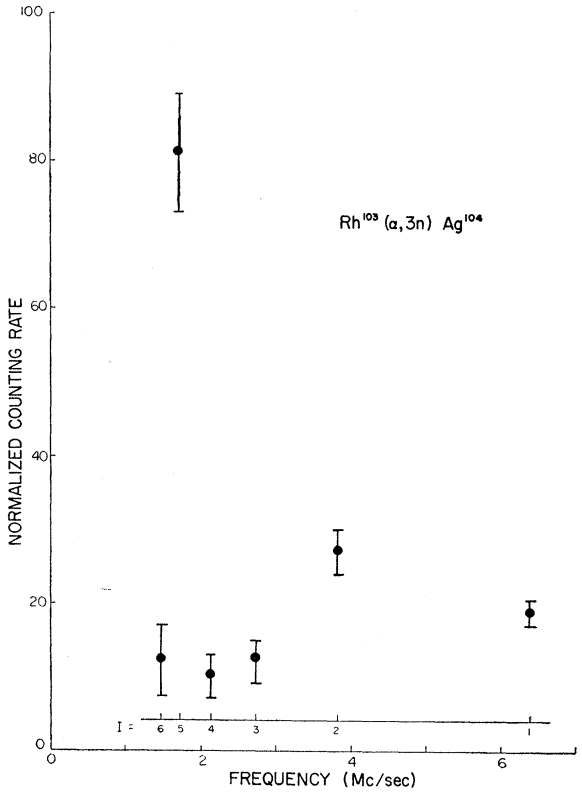

FIG. 6. Spin search for 27-min Ag^{104} at 6.8 gauss. The strong signal on $I=5$ is due to ~1-hr Ag^{104}. The exposure for $I=1$ appears higher than it should be (see Table I).

TABLE I. Analysis of spin exposures confirming the resonance of 27-min Ag^{104} on spin 2.

Exposure	27-min Ag^{104} (counts/min)	1.2-hr Ag^{104} (counts/min)
$I=1$	0 (3)	33 (2)
$I=2$	30 (4)	35 (2)
$I=3$	6 (3)	34 (2)
"Half-beam"	207 (17)	1113 (9)

the $(\alpha,4n)$ reaction with the available energy of $\sim$45 Mev (the aluminum cover foil for the illustrated experiment was only one mil thick). The $I=5$ resonance for 1.2-hr Ag^{104} has been observed several times at frequencies from 1.4 Mc/sec to 20 Mc/sec.

The detection and identification of the spin resonance of 27-min Ag^{104} is made difficult by the unavoidable presence of the 1.2-hr isomer. Furthermore, the counting efficiency of the crystal x-ray counters is sharply reduced for the predominantly β^+ decay[10] of the shorter isomer (see Fig. 1). As a consequence, the spin search shown in Fig. 6 is not so definitive for the 27-min isomer as might be desired, although the spin 5 for 1.2-hr Ag^{104} is quite strong. A check of the decay curves for the possible spin resonance on spin 1 or 2 showed that the decay of the spin 2 sample was more rapid than either the spin 1 sample or a half-beam exposure taken at about the same time. To check this, exposures were made for $I=1$, 2, 3 and counted in continuous-flow gas β counters. An analysis of these three samples gave the results listed in Table I. For the sake of comparison, the beam composition is shown by an analysis of a half-beam exposure. Even using β counters, the background of 1.2-hr activity is seen to be very large. However, the resonance on spin 2 of 27-min Ag^{104} is evident. This spin 2 resonance has been observed at six frequencies between 3.8 Mc/sec and 6.6 Mc/sec.

The $\sim$1-hr activity reported by Reynolds *et al.*[5] as having spin 2 has not been observed. If the activity were to have a mass number $\geqslant 103$, it should have appeared in at least one of the above spin searches.

DISCUSSION

The high-spin–low-spin isomerism observed for these isotopes is not unusual in this region of the periodic table. The principle reason is the competition between $g_{9/2}$ and $p_{\frac{1}{2}}$ proton levels from $Z=38$ to $Z=50$.[11] For the isotopes reported here, both N and Z are near the magic number 50, so that the spherical shell model is expected to provide a good approximation to the nuclear states. Using the group approach suggested by Way *et al.*,[12] the possible neutron and proton levels can be combined by Nordheim's rules[13] in a limited number of ways. These are summarized in Table II. An italicized number indicates that the spin is predicted by an application of the strong rule, while the absence of italics indicates a possible spin predicted by the weak rule. Solely on the basis of atomic-beam measurements (reference 5 and this paper), the configurations of the isomers of silver-104 and silver-106 are uniquely determined by the assumption of group validity of Nordheim's rules.[12]

TABLE II. Possible spin assignments for the odd-odd isotopes of silver.

Neutron \ Proton	$p_{1/2}$	$g_{9/2}$
$d_{5/2}$	2^-	3^+, 4^+, 5^+, 6^+, 7^+
$g_{7/2}$	4^-	1^+

The results for Ag^{106} have been anticipated by Bendel *et al.*[14] who, from decay-scheme systematics, assigned a state of 1^+ to the 24-min isomer and a spin $\geqslant 4$ for the 8.3-day isomer. The present value of 6 for the spin of the long-lived isomer leads to the assignment of even parity to the nuclear state, on the basis of the simple shell model.

In the case of Ag^{104}, the spin of the short-lived isomer was assigned by Johnson.[10] However, the parity of the state has been predicted by him to be even, which is in conflict with the shell-model picture presented in Table II.

The 1.2-hr Ag^{104} has not been previously observed in detail. Several authors,[9,10,14–16] report the presence of an $\sim$1-hr silver activity in various experiments, but mass assignments have been uncertain. Haldar and Wiig[16] definitely assigned a 1.1-hr half-life to Ag^{103} on the basis of a decay to 17-day Pd^{103}. The 59-min half-life which Johnson[10] assigns to Ag^{103} may be expected to be partly due to Cd^{104} and Cd^{105}, as he suggests.

It is apparent, in view of the present experiments, that there are in fact at least two silver isotopes with half-lives of $\sim$1 hr. One of these must be Ag^{103} with $I=\frac{7}{2}$; another must be Ag^{104} with $I=5$.

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